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Atmospheric Processes***

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ASSESSING THE MERCURY HEALTH RISKS ASSOCIATED WITH COAL-FIRED POWER PLANTS: ISSUES IN ATMOSPHERIC PROCESSES

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Introduction

The rationale for regulating air emissions of mercury from U.S. coal-fired power plants largely depends on mathematical dispersion modeling, including the atmospheric chemistry processes that affect the partitioning of Hg emissions into elemental (Hg_0) and the reactive (RGM) forms that may deposit more rapidly near sources. This paper considers and evaluates the empirical support for this paradigm. We consider the extant experimental data at three spatial scales: local (< 30 km), regional (< ~300 km), and national (multi-state data). An additional issue involves the finding of excess Hg levels in urban areas.

Local Evidence

Our search of the literature found seven studies dating back 30 years in which various manifestations of local Hg deposition were related to coal-fired power plants (Table 1). These experiments involved concentrations of Hg in soil, lake sediments, precipitation and fish. To place these results in a common framework, we estimated the relationships between background concentrations and those obtained near the plant and the fractions of emitted Hg that had been deposited and retained during the period of facility operation. Where possible, we accounted for these relationships as a function of the receptor's distance from the plant.

Soil Composition. The earliest attempts at assessing Hg impacts from coal-fired power plants were based on the Hg content of surficial soil samples¹⁻³. This technique is highly dependent upon definition of the background soil content, which was not always reliable. The reported incremental Hg concentrations ranged from 29%¹ to 42%² above background, corresponding to retention of 3-5% of cumulative emissions. The Four Corners study³ did not measure background but concluded that "mercury was not accumulating in the soil", based mainly on comparisons of local soil Hg concentrations with those reported in the literature for other locations. However, reanalysis of the published data on individual soil samples shows a significant overall (log-log) slope of -0.11 ($p < 0.005$) as a function of downwind distance. Moreover, scatter plots showed wide variation by wind direction, a more rapid decrease in soil Hg near the plant, and the suggestion of a secondary peak at about 10-15 km downwind. A possible rationale might thus be a close-in peak due to rainout and a more distant peak due to plume touchdown and dry deposition. We assumed various values of background Hg in order to estimate the fraction of emissions deposited and retained in the soil; the wide range of these estimates shows the sensitivity to this parameter. It appears that around 10% of the plant's Hg emissions may have been deposited and retained in the soil, which would correspond to excess deposition rates of about 60%. Such estimates are also sensitive to the maximum downwind distance considered, which was 20-30 km in these various examples.

Sediments. Three studies^{2,5,6} include data on the Hg content of sediment cores obtained from nearby water bodies. This technique has the advantage of potential relevance to impacts on fish but has the disadvantages of uncertain time periods and the difficulty of

considering decay rates in terms of downwind distance. At the Kincaid plant², the excess Hg in sediments was reported to be about 30%. In Texas⁶, there was about 18% more Hg in sediments from a lake near the plant than from two lakes about 30 km away. The decreases in Hg with depth within the core were also substantially larger in the sediment cores from the nearby lake.

Precipitation. Two studies^{5,7} considered variations in the Hg content of precipitation as a marker for local impact. In Slovenia⁵, it was not clear that valid annual averages had been obtained and no comparisons were made with local background. In Indiana⁷, the authors reported no significant difference between the precipitation Hg data collected near the power plant and that collected at 3 other sites in the state. However, precipitation chemistry is frequently sensitive to the rate and thus amount of precipitation, and when this factor is taken into account in a multiple regression analysis, the site near the plant appears to have an excess Hg level of about 12% (Figure 1). With normalized deposition (deposition/precipitation) as the dependent variable, the increment is just significant ($p < 0.03$).

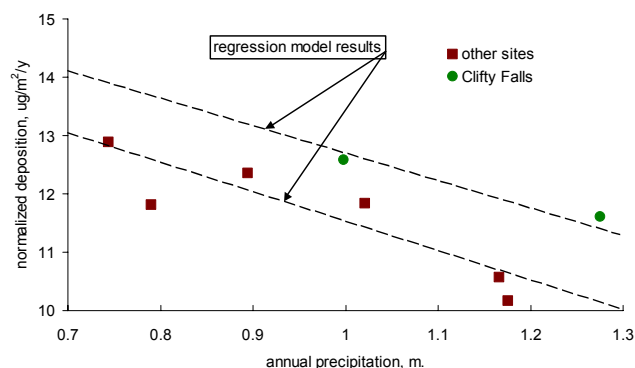


Figure 1. Comparison of wet deposition at Clifty Falls Mercury Deposition Network (MDN) monitoring site.

Fish Hg Content. Two studies^{2,4} reported the Hg content found in various species of fish collected in or near the impacts zones of power plants. At Kincaid², the fish caught near the plant had substantially less Hg than those from more distant locations, but it was not clear whether this comparison included adjustments for fish size. For each of the 23 farm ponds in the Dickerson study⁴, fish lengths and detailed water chemistry data were also reported, which turned out to be important. To examine these factors, we estimated wet deposition of Hg to each pond based on the modeled isopleth plots in the paper. In multiple regression analysis, the log of fish Hg content was significantly ($p < 0.01$) associated with the log of wet Hg deposition (coefficient = 0.5), pond water conductivity (coefficient = -0.5), and fish length for sunfish and bass pooled ($n=37$). Fish length was essentially a surrogate for fish species. Water conductivity, hardness, and alkalinity were highly inter-correlated; pH was not a significant predictor of fish Hg. Water quality was not associated with estimated Hg wet deposition. Figure 2 is scatter plot of these data, based on fish Hg adjusted to a common level of pond water conductivity. Note that only one sample clearly exceeds the EPA guideline for fish Hg content (0.3 ppm), notwithstanding the effects of the power plant; 3 samples exceeded this level in the unadjusted data.

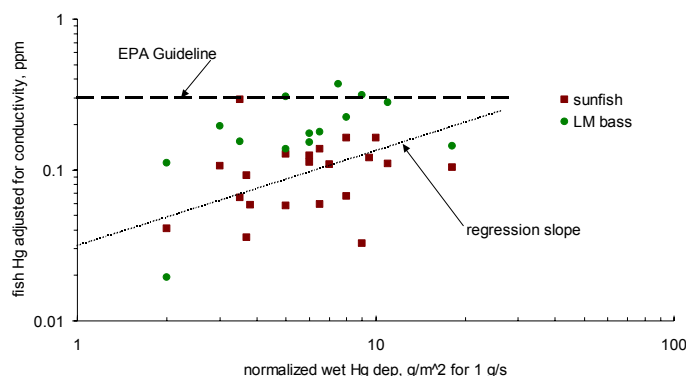


Figure 2. Comparison of measured fish Hg (ppm) as a function of predicted wet deposition.

Regional Data

As an example of regional relationships, we used data on Hg emissions from power plants⁸ and on 1998-2002 wet Hg deposition obtained from the Mercury Deposition Network (MDN)⁹, both for Pennsylvania. The 8 MDN stations operated for various portions of this period; each annual wet deposition estimate was thus treated as a separate observation ($n=22$). The straight-line distance between each MDN site and each of the 36 plants, whose Hg emissions ranged from 637 kg to 45 g, totaling 3622 kg, was computed from latitude-longitude data, and each emission rate was weighted by the inverse square of this distance and summed. For the 8 MDN sites, these sums ranged from 0.09 kg/km² to 1.7 kg/km².

Multiple regression analysis was then used to deduce source-receptor relationships, considering the year, site elevation, and site latitude as possible confounders. No account was taken of other point sources of Hg (such as urban areas or out-of-state sources) or of prevailing wind directions for precipitation events. MDN site latitude was included as a possible confounder because the national map of Hg wet deposition seems to show consistent increases from north to south in the eastern states.

The coefficients for site elevation and year were far from significant and were dropped from further consideration. Latitude and (emissions/distance²) were moderately (negatively) correlated, which posed a co-linearity problem. However the best fit to these 22 observations was obtained in a log-log regression based on the emission parameter alone, which had a log-log coefficient of 0.11 ($p<0.010$). This result implies that 11% of the wet Hg deposition in Pennsylvania is associated with coal-fired power plants in the state, under the modeling assumptions stated above. It also implies that wet deposition of Hg may decrease more slowly with downwind deposition than originally assumed. However, when various assumed levels of background wet deposition are subtracted from the measured MDN values, the regression coefficients are increased, at the expense of poorer model fits.

As an example, this empirical model would predict the following annual deposition ($\mu\text{g}/\text{m}^2$)-distance relationship for an isolated point source of 636 kg Hg per year: 1 km., 19.5; 3 km, 15.3; 10 km, 11.7, 30 km, 9.2; 100 km, 7.1. Taking the last figure as "background", the annual wet deposition would be about 1.1% of emissions and the total Hg deposition, about 2.2%.

Additional information on Hg deposition at the regional scale is provided by the results of Lopez-Alonzo et al.¹⁰, who analyzed the Hg content of calf kidneys in relation to distances between the farms and major point sources of Hg in Northwest Spain. (e.g. 1200 kg/y from coal-fired power plants and similar amounts from other

industries). Downwind distances ranged up to 140-200 km; the duration of this deposition was not mentioned. Based on approximations from the scatter-plots¹⁰, it appears that deposition decreased with downwind distance to the 0.4 power for the power plants and 0.7 power for the industrial area. Neither of these slopes is significantly different from the expected square-root relationship.

The National Scale

We used state-level data on Hg deposition (MDN) and fish Hg concentrations¹¹ to deduce larger scale source-receptor relationships. The deposition data were averaged over time (1997-2002) and the fish Hg data were considered by species (9 different species). A simple regression model was used in which $\ln(\text{fish Hg})$ was regressed against $\ln(\text{deposition})$ and dummy variables for fish species. This model assumes that all species react to deposition in the same way and provides fish Hg concentration increments relative to a referent species, in this case large-mouth bass. This approach provided 105 combinations of states and fish species (only 31 states were represented). Channel catfish, bluegills, common carp, white suckers, and yellow perch all had significantly lower Hg concentrations than bass, walleye, or northern pike (as expected). However, the effect of Hg deposition, as averaged over entire states, was significantly negative in this model ($p < 0.025$). Latitude was not an important confounder when both eastern and western states were considered. When large-mouth bass were considered alone ($n=20$), the relationship with deposition was positive but far from significance (log-log coefficient = 0.21).

A further consideration at the national scale is provided by the recent global modeling study of Seigneur et al.¹², who estimated that 25-32% of the total Hg deposition to the contiguous US came from North American anthropogenic sources. These sources total 200.1 metric tons/y, of which electric utilities comprise 52.7 tons, or 26.3%. Assuming proportionality, the utility share of deposition would then be about 6.6-8.4%, which is in line with the various estimates above that are based on actual measurements. Comparison of these figures suggests a minor role for Hg deposition at distances $> \sim 100$ km.

Discussion

The above findings lead to the overall conclusion that atmospheric deposition of Hg is affected by emissions from coal-fired power plants. However, because of the numerous assumptions required and the use of simplistic models, it is not possible to accurately describe these relationships on this basis. Complex atmospheric chemistry and dispersion models are required to predict precise concentration and deposition contributions, and aquatic process models are required to predict effects on fish.

These caveats notwithstanding, it is still useful to summarize the consistencies and inconsistencies in our findings. In terms of excesses over background, we see local soil concentration Hg increments of 30%-60%, sediment increments of 18-30%, and wet deposition increments of 11-12%. If we accept the finding⁷ that fish Hg is proportional to the square root of wet deposition (after controlling for water chemistry), then the contribution of coal-fired power plants to fish Hg would be about 5-6%. It is possible that the absence of local impacts on fish at Kincaid was due to water chemistry. Local differences in water chemistry may also help explain the absence of a relationship between state-level fish concentrations and wet deposition levels.

Effects on public health should be less than a few percent as more than 90% of the population currently meets the EPA exposure guideline and few individuals consume large quantities of fresh-water fish where coal is burned.

Finally, it should be noted that none of these simple analyses have accounted for possible impacts from urban areas, as implied by recent findings of excess urban deposition.^{13,14} A mass-balance study of Hg deposition to Lake Michigan¹⁵ showed a slow rate of decrease in deposition (in terms of distance from Chicago), as did the data on power plants discussed above.

Given the resources required to control mercury from power plants, a comprehensive campaign to measure their mercury impacts in detail and to monitor any ensuing changes would be prudent.

Acknowledgment

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Table 1 Local Hg deposition from coal-fired power plants

reference	plant data	emissions	deposition data, ppb in soils, ug/m ² /yr in precip				percent		remarks
		kg/y	type	# samples	mean	background	deposited		
Klein & Russell (1973)	Campbell, MI (?) 650 MW 122 m stack	~290	soil	90	10.2	7.9	2.7		irregular impact area
Anderson & Smith (1977)	Kincaid (IL) 1200 MW 2 152 m stacks	531	soil	90	22	15.5	4.6		limited to 1 sector of the impact area ~1% of emissions deposited in lake fish near plant unaffected
			sediment	36	49	37			
			fish	120	70-82	110-560			
Crockett & Kinnison (1979)	Four Corners (NM) 2150 MW 2 76, 2 91 m stacks	595	soil	70	14.5	*0	20.6		*assumed background values
						*3	16.3		
						*6	12.0		
						*9	7.8		
Pinkney et al. (1997)	Dickerson (MD) 543 MW	~240	fish	69 sunfish 42 LM bass					fish Hg in 23 small farm ponds increased with (Hg dep) ^{0.5}
Kotnik et al. (2000)	Sostanj, Slovenia 775 MW 100, 150, 230 m stacks	314	precip lake sediments		7.4-13.7 ug/m2 53-166		5		based on 20 km radius and dry dep = wet higher values in surface layers
Menounou & Presley (2003)	Gibbons Cr (TX) 460 MW	~430	lake sediments	13 (cores)	94 (data from top layers)	80	30		based on 25 km radius and median excess deposit of 0.094 ug/cm2
Risch (2003)	Clifty Cr (IN) 1300 MW	184	precip	4 sites, 2 yrs	12.3	11			ug/m2/y at 110 cm precip (monitor 3 km from plant) Other plants in Ohio and Kentucky not considered